

Preparation and characterization of a composite PDMS membrane on CA support

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Polydimethylsiloxane (PDMS) is the most commonly used membrane material for the separation of condensable vapors from lighter gases. In this study, a composite PDMS membrane was prepared and its gas permeation properties were investigated at various upstream pressures. A microporous cellulose acetate (CA) support was initially prepared and characterized. Then, PDMS solution, containing crosslinker and catalyst, was cast over the support. Sorption and permeation of C₃H₈, CO₂, CH₄, and H₂ in the prepared composite membrane were measured. Using sorption and permeation data of gases, diffusion coefficients were calculated based on solution-diffusion mechanism. Similar to other rubbery membranes, the prepared PDMS membrane advantageously exhibited less resistance to permeation of heavier gases, such as C₃H₈, compared to the lighter ones, such as CO₂, CH₄, and H₂. This result was attributed to the very high solubility of larger gas molecules in the hydrocarbon-based PDMS membrane in spite of their lower diffusion coefficients relative to smaller molecules. Increasing feed pressure increased permeability, solubility, and diffusion coefficients of the heavier gases while decreased those of the lighter ones. At constant temperature (25°C), empirical linear relations were proposed for permeability, solubility, and diffusion coefficients as a function of transmembrane pressure. C₃H₈/gas solubility, diffusivity, and overall selectivities were found to increase with increasing feed pressure. Ideal selectivity values of 9, 30, and 82 for C₃H₈ over CO₂, CH₄, and H₂, respectively, at an upstream pressure of 8 atm, confirmed the outstanding separation performance of the prepared membrane. Copyright © 2009 John Wiley & Sons, Ltd.

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INTRODUCTION

The separation of higher hydrocarbons, such as C₃H₈, from lower ones, like CH₄, in a flue or flare gas in a refinery, is of great economic importance. As the rates of these gas streams are usually modest, applying membrane gas separation process seems to be rational.

The most important part of a membrane separation process is the membrane itself, and polymeric membranes are the most common ones used in membrane gas separation process. Gas permeability through dense polymeric membranes is typically calculated by multiplying diffusion and solubility coefficients of the penetrants. In all polymer materials, diffusion coefficient decreases with increasing molecular size. It is due to the fact that large molecules interact with more segments of the polymer chains than the smaller molecules, thereby favoring the passage of small molecules, such as H₂, over larger ones such as C₃H₈. However, solubility increases with increasing condensability and therefore increases with increasing molecular size.

In glassy, rigid polymers, such as polysulfone (PS), permeant diffusion coefficient is more important than solubility coefficient. Therefore, these polymers preferentially permeate the smaller, noncondensable gases, H₂ and CH₄, over the larger condensable gases, C₃H₈ and CO₂. On the other hand, in rubbery polymers, such as polydimethylsiloxane (PDMS), permeant solubility coefficient is the most important. Thus, these polymers preferentially permeate the larger, more condensable gases over the smaller,

noncondensable gases.^[1] Higher hydrocarbons are usually the minor components of flue or flare gas streams in a refinery.^[2] Hence, when rubbery membranes, such as PDMS, are used, a small portion of feed has to permeate through the membrane to remove from the feed stream, requiring relatively small membrane areas.

PDMS is the most commonly used rubbery membrane material for separation of higher hydrocarbons from permanent gases. Recently, many studies have been carried out on transport properties of pure and binary gas mixtures of O₂, N₂, H₂, CO, CO₂, CH₄, and C₂–C₄ olefins and paraffins using PDMS membrane.^[3–25] PDMS membranes, which have been prepared heretofore and evaluated in gas separation applications, were in the form of a single layer,^[3–8] a composite with a microporous support,^[9–20] a mixed matrix,^[21–23] and a copolymer.^[24,25] Most of the composite

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